AMENDMENT

(Amendment under of Article 11 of the Japanese Law [PCT Article 34(2)(b)])

To: Commissioner of the Patent Office

1. Identification of the International Application

PCT/JP03/016542

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4. Items to be Amended

Description and Claim

5. Contents of Amendment

(1) "As one example - - (see the following Patent Publication 15)" is added between lines 6 and 7, page 3 of the specification.

(2) " Patent Publication 15: Japanese Patent Laid-Open Publication No. 2001-338808" is added between lines 20 and 21, page 3 of the specification.

(3) "a spherical particle of metallic glass alloy prepared by an atomizing process" on the lines 16 to 17, page 6 of the specification is amended to "a spherical particle for use in producing a bulk Fe-based sintered alloy soft magnetic material of metallic glass, comprising a Fe-based metallic

glass alloy prepared by an atomizing process".

(4) " prepared by sintering the plurality of spherical particles of metallic glass alloy set forth in the first aspect of the present invention" on line 22, page 6 of the specification is amended to " prepared by sintering the plurality of spherical particles of Fe-based metallic glass alloy set forth in the first aspect of the present invention at a temperature of 573 K or more".

(5) "obtain an alloy particle" on line 7, page 8 of the specification is amended to "obtain a Fe-based metallic glass alloy particle".

(6) "a sintering temperature (T) is set in a temperature range" on line 14, page 8 of the specification is amended to "a sintering temperature (T) is set at 573 K or more and within a temperature range".

(7) "Fe-based metallic glass alloy prepared by an atomizing process" in claim 1 is amended to "comprising a Fe-based metallic glass alloy prepared by an atomizing process".

(8) " A spherical particle" in claim 1 is amended to "A spherical particle for use in producing a bulk Fe-based sintered alloy soft magnetic material of metallic glass".

(9) "Fe-based metallic glass alloy as defined in claim 1" in claim 1 is amended to "Fe-based metallic glass alloy as defined in claim 1 at a temperature of 573 K or more".

(10) "obtain an alloy particle" in claim 4 is amended to "obtain a Fe-based metallic glass alloy particle".

(11) "a sintering temperature (T) is set in a temperature range" in claim 5 is amended to "a sintering temperature (T) is set at 573 K or more and within a temperature range".

sintered body prepared by sintering plate-shaped particles of Fe-based (Fe-Al-Ga-P-C-B-Si based, etc.) amorphous alloy in a temperature range of 693 to 713 K (see the following Patent Publication 14). Further, the inventors reported a Fe-based soft magnetic metallic glass sintered body prepared by spark-discharging particles obtained through a gas atomizing process, which have a particle size of 10 to 30 μm, and a primary component of Fe-Co-Ga-P-C-B based amorphous alloy (see the following Non-Patent Publications 1 to 3).

As one example other than a production method for a metallic glass sintered body, there has also been known a method for producing a powder magnetic core, which comprises, adding 0.2 to 5 wt% of insulating material, such as epoxy resin, silicone resin or water glass, to a Fe-Al-Ga-P-C-Bi-Si based or Fe-Ga-P-C-Bi-Si based metal glass powder having a particle size of 30 to 300 μm, mixing them, subjecting the obtained mixture to a compression molding process in a temperature range of 373 to 573 K to moderately soften the insulating material so as to bind the metallic glass alloy powder together to form the mixture in a given shape (see the following Patent Publication 15)

Patent Publication 1: Japanese Patent Laid-Open Publication No. 08-333660

Patent Publication 2: Japanese Patent Laid-Open Publication No. 09-320827

Patent Publication 3: Japanese Patent Laid-Open Publication No. 11-071647

Patent Publication 4: Japanese Patent Laid-Open Publication No. 2001-152301

Patent Publication 5: Japanese Patent Laid-Open Publication No. 2001-316782

Patent Publication 6: Japanese Patent Laid-Open Publication No. 2002-226956

Patent Publication 7: Japanese Patent Laid-Open Publication No. 11-073608

Patent Publication 8: Japanese Patent Laid-Open Publication No. 11-073609

Patent Publication 9: Japanese Patent Laid-Open Publication No. 11-074109

Patent Publication 10: Japanese Patent Laid-Open Publication No. 11-074111

Patent Publication 11: Japanese Patent Laid-Open Publication No. 08-337839

Patent Publication 12: Japanese Patent Laid-Open Publication No. 10-092619

Patent Publication 13: Japanese Patent Laid-Open Publication No. 11-071648

Patent Publication 14: Japanese Patent Laid-Open Publication No. 2000-345308

Patent Publication 15: Japanese Patent Laid-Open Publication No. 2001-338808

Non-Patent Publication 1: Baolong Shen et al., "Bulk Formation by Spark-Plasma Sintering of Fe-Co-Ga-P-C-B Glass Alloy Powder and Magnetic Characteristics thereof", Powder and Powder Metallurgy, Vol. 48, No. 9, September 2001, pp. 858-862

Non-Patent Publication 2: Baolong Shen et al., "Preparation of $Fe_{65}Co_{10}Ga_5P_{12}C_4B_4$ Glassy Alloy with Good Soft Magnetic Properties by Spark-Plasma Sintering of Glassy Power", Materials Transactions, Vol. 43, No. 8, p. 1961-1965 (2002)

Non-Patent Publication 2: Baolong Shen et al., "Preparation of $Fe_{65}Co_{10}Ga_5P_{12}C_4B_4$ Metallic Glass Magnetic Core by Spark-Plasma Sintering", "Journal of Japan Society of Powder and Powder Metallurgy", November 2002, p. 196

magnetic characteristics to an atomizing process having a low cooling rate so as to obtain a spherical metallic glass alloy particle with a large particle size, and to subject the plurality of spherical metallic glass alloy particles to a spark plasma sintering process under a high compression pressure so as to prepare a high-density sintered body consisting of a metallic glass phase having a relative density of 99.0 % or more, or provide a bulk Fe-based sintered alloy soft magnetic material of metallic glass having extremely excellent soft magnetic characteristics.

A metallic glass for use in producing an amorphous soft magnetic alloy sintered body of the present invention has a temperature interval of a supercooled liquid region (ΔTx) of 25 K or more, preferably 40 K or more, as expressed by the following formula: $\Delta Tx = Tx - Tg$ (wherein Tx is a crystallization (onset) temperature, and Tg is a glass transition temperature), and a reduced glass transition temperature of 0.59 or more, as expressed by the following formula: Tg / Tl (wherein Tg is a glass transition temperature, and Tl is a liquidus temperature). These characteristics make it possible to readily produce an alloy particle consisting of a single phase of metallic glass and having an approximately complete spherical shape, through a high-pressure-gas atomizing process.

Specifically, according to a first aspect of the present invention, there is provided a spherical particle for use in producing a bulk Fe-based sintered alloy soft magnetic material of metallic glass, comprising a Fe-based metallic glass alloy prepared by an atomizing process, which has a particle size of 30 to 125 μm, and a composition consisting of, by atomic %, 0.5 to 10 % of Ga, 7 to 15 % of P, 3 to 7 % of C, 3 to 7 % of B and 1 to 7 % of Si, with the remainder being Fe.

According to a second aspect of the present invention, there is provided a bulk Fe-based sintered alloy soft magnetic material of metallic glass, which consists of a metallic glass phase high-density sintered body with a relative density of 99.0 % or more, prepared by sintering the plurality of spherical particles of Fe-based metallic glass alloy set forth in the first aspect of the present invention at a temperature of 573 K or more, and has a magnetic permeability of 3900 (μmax) or more and a coercive force (Hc) of 19 (A/m) or less in an as-sintered state. The metallic glass has a temperature interval of a supercooled liquid region (ΔTx) of 25 K or more,

as expressed by the following formula: $\Delta Tx = Tx - Tg$ (wherein Tx is a crystallization temperature, and Tg is a glass transition temperature), and a reduced glass transition temperature of 0.59 or more, as expressed by the following formula: Tg / Tl (wherein Tg is a glass transition temperature, and Tl is a liquidus temperature).

7000 (μmax) or more and a coercive force (Hc) of 12 (A/m) or less.

According to a fourth aspect of the present invention, there is provided a method of producing the spherical particle set forth in the first aspect of the present invention, which comprises preparing molten alloy having a composition consisting of, by atomic %, 0.5 to 10 % of Ga, 7 to 15 % of P, 3 to 7 % of C, 3 to 7 % of B and 1 to 7 % of Si, with the remainder being Fe, dropping or ejecting the molten alloy from a nozzle, and spraying high-speed gas to droplets of the molten alloy to rapidly solidify the droplets so as to obtain a Fe-based metallic glass alloy particle having an amorphous phase and a maximum particle size of 30 to 125 μm.

According to a fifth aspect of the present invention, there is provided a method of producing the Fe-based sintered alloy soft magnetic material set forth in the second aspect of the present invention, which comprises preparing a plurality of spherical particles of Fe-based metallic glass alloy having a particle size of 30 to 125 μm by the method set forth in the fourth aspect of the present invention, and sintering the spherical particles by a spark plasma sintering process under the conditions that: a heating rate is set at 40 K/min or more; a sintering temperature (T) is set at 573 K or more and within a temperature range satisfying a relationship of $T \leq Tx$, wherein Tx is a crystallization temperature; and a sintering pressure is set at 200 MPa or more.

According to a sixth aspect of the present invention, there is provided a method of producing the bulk Fe-based sintered alloy soft magnetic material of metallic glass set forth in the third aspect of the present invention, which comprises preparing a Fe-based sintered alloy soft magnetic material by the method set forth in the fifth aspect of the present invention, and subjecting the Fe-based sintered alloy soft magnetic material to a heat treatment in a temperature range of 573 to 723 K.

The Fe-based sintered alloy soft magnetic material of the present invention has a soft magnetism at room temperature, and exhibits a high saturation magnetization of 1.3 to 1.4 T. Further, the Fe-based sintered alloy soft magnetic material has a Curie temperature of 600 K or more, and thereby has a thermal stability in the magnetic characteristics. This sintered body exhibits a high specific resistance value of 1.6 μΩm or more.

Each value of the above characteristics was measured from a sample prepared by sintering the spherical particles in a disc shape having a diameter of 20 mm and a thickness of 5 mm using

What is claimed is:

1. (Amended) A spherical particle for use in producing a bulk Fe-based sintered alloy soft magnetic material of metallic glass, comprising a Fe-based metallic glass alloy prepared by an atomizing process, which has a particle size of 30 to 125 μm, and a composition consisting of, by atomic %, 0.5 to 10 % of Ga, 7 to 15 % of P, 3 to 7 % of C, 3 to 7 % of B and 1 to 7 % of Si, with the remainder being Fe.

2. (Amended) A bulk Fe-based sintered alloy soft magnetic material of metallic glass, which consists of a metallic glass phase high-density sintered body with a relative density of 99.0 % or more, prepared by sintering the plurality of spherical particles of Fe-based metallic glass alloy as defined in claim 1 at a temperature of 573 K or more, and has a magnetic permeability of 3900 (μmax) or more and a coercive force (Hc) of 19 (A/m) or less in an as-sintered state, wherein said metallic glass has:

a temperature interval of a supercooled liquid region (ΔTx) of 25 K or more, as expressed by the following formula: $\Delta Tx = Tx - Tg$, wherein Tx is a crystallization temperature, and Tg is a glass transition temperature; and

a reduced glass transition temperature of 0.59 or more, as expressed by the following formula: Tg / Tl, wherein Tg is a glass transition temperature, and Tl is a liquidus temperature.

3. A bulk Fe-based sintered alloy soft magnetic material of metallic glass, prepared by subjecting the bulk Fe-based sintered alloy soft magnetic material as defined in claim 2 to a heat treatment in a temperature range of 573 to 723 K, which has a magnetic permeability of 7000 (μmax) or more and a coercive force (Hc) of 12 (A/m) or less.

4. A method of producing the spherical particle as defined in claim 1, comprising:

preparing molten alloy having a composition consisting of, by atomic %, 0.5 to 10 % of Ga, 7 to 15 % of P, 3 to 7 % of C, 3 to 7 % of B and 1 to 7 % of Si, with the remainder being Fe;

dropping or ejecting said molten alloy from a nozzle; and

spraying high-speed gas to droplets of said molten alloy to rapidly solidify said droplets so as to obtain a Fe-based metallic glass alloy particle having an amorphous phase and a maximum particle size of 30 to 125 μm.

5. (Amended) A method of producing the Fe-based sintered alloy soft magnetic material as defined in claim 2, comprising:

preparing a plurality of spherical particles of Fe-based metallic glass alloy having a particle size of 30 to 125 μm by the method as defined in claim 4; and

sintering said spherical particles by a spark plasma sintering process under the conditions that: a heating rate is set at 40 K/min or more; a sintering temperature (T) is set at 573 K or more and within a temperature range satisfying a relationship of $T \leq Tx$, wherein Tx is a crystallization temperature; and a sintering pressure is set at 200 MPa or more.

6. A method of producing the bulk Fe-based sintered alloy soft magnetic material of metallic glass as defined in claim 3, comprising:

preparing a Fe-based sintered alloy soft magnetic material by the method as defined in claim 5; and

subjecting said Fe-based sintered alloy soft magnetic material to a heat treatment in a temperature range of 573 to 723 K.